Observation of the underscreened Kondo effect in a molecular transistor

Nicolas Roch,[1] Serge Florens,[1] Theo A. Costi,[2] Wolfgang Wernsdorfer,[1] and Franck Balestro[1]

[1] *Institut Néel, associé à l'UJF, CNRS, BP 166, 38042 Grenoble Cedex 9, France*

[2] *Institut für Festkörperforschung, Forschungszentrum Jülich, 52425 Jülich, Germany*

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We present first quantitative experimental evidence for the underscreened Kondo effect, an uncomplete compensation of a quantized magnetic moment by conduction electrons, as originally proposed by Nozières and Blandin. The device consists of an even charge spin $S = 1$ molecular quantum dot, obtained by electromigration of C_{60} molecules into gold nanogaps and operated in a dilution fridge. The persistence of logarithmic singularities in the low temperature conductance is demonstrated by a comparison to the fully screened configuration obtained in odd charge spin $S = 1/2$ Coulomb diamonds. We also discover an extreme sensitivity of the underscreened Kondo resonance to magnetic field, that we confirm on the basis of numerical renormalization group calculations.

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When a magnetic impurity is inserted in a piece of metal, its magnetic moment can be completely screened by the conduction electrons, owing to their quantized spin 1/2. This general phenomenon, the Kondo effect, has been thoroughly studied in diluted magnetic alloys [1] and has attracted considerable attention in the more recent quantum dot systems [2]. Clearly, impurities carrying a spin S greater that 1/2 need to bind several electronic orbitals in order to fully quench their magnetism, and Nature seems to conspire in always providing enough screening channels for that situation to occur in general [3]. Therefore, the possibility that screening may happen to be incomplete, as initially proposed on theoretical grounds by Nozières and Blandin [4], has remained elusive for almost thirty years, despite the great experimental control that one can achieve with artificial quantum dot setups. The observation of the underscreened Kondo effect, in addition to its overscreened counterpart [5], is also especially appealing since it constitutes one of the simplest cases where standard Fermi Liquid Theory is violated [6, 7].

We demonstrate in this Letter that molecular quantum dots obtained through electromigration [8] are perfect candidates for achieving underscreened Kondo impurities. Indeed point contact tunneling (single mode) and important left/right asymmetry of the transport electrodes ensure a large window of energies where a *single* screening channel is active. In addition, the Kondo phenomenon in molecules can set in already at several kelvins [9, 10, 11, 12] thanks to relatively important charging energies, allowing a complete study of Kondo crossovers on a sufficient range of temperatures. Both conditions of single channel and large Kondo temperature are difficult to meet altogether in other quantum dot devices, where Kondo effects associated with higher spin states have been previously found, but yet not investigated in detail [13, 14, 15, 16, 17]. We report here on the first observation of the anomalous logarithmic behavior in the temperature and bias voltage dependent conductance in a spin $S = 1$ quantum dot below the Kondo scale, as previously predicted for underscreened impurities [4, 6, 7, 18], and successfully confront our results with quantitative numerical renormalization group (NRG) calculations. More strikingly, the experimental data demonstrate that underscreened impurities are extremely sensitive to the application of a magnetic field. In that case, and in contrast to fully screened moments, the Kondo resonance is split by a Zeeman energy much smaller than the Kondo temperature, reflecting the high degree of polarizability of a partially screened spin. This surprising finding is confirmed by new NRG calculations of the local density of states that we performed on the single-channel spin $S = 1$ Kondo model in a magnetic field.

Recent work by us [19] has shown that C_{60} quantum dots with two electrons on the molecule can be gate-tuned via a quantum phase transition between a molecular singlet and a spin $S = 1$ Kondo state. Interestingly, the central condition for this phase transition to occur, vindicated by these experimental findings [19], is the presence of a single screening channel in the accessible temperature range [20, 21]. Since screening channels result from the overlap between the wave functions of the conduction electrons and those of the magnetic impurity orbitals [4], one understands that orbital quantum numbers must not be preserved in order for the underscreened Kondo effect to take place, see Fig.1.a discussing the situation of $n_{\rm sc}$ channels coupled to an impurity spin S. We can describe our molecular transistor [19] by two orbital levels $(-1, +1)$ coupled to two metallic leads (L, R) (see figure 1.b), with the tunneling matrix [20]:

$$\mathbf{t} = \begin{pmatrix} t_{L,+1} & t_{R,+1} \\ t_{L,-1} & t_{R,-1} \end{pmatrix} \tag{1}$$

A screening channel $\lambda = 1, 2$ is associated to each eigenvalue t_λ of this matrix, from which result antiferromagnetic Kondo couplings $J_\lambda = 8|t_\lambda|^2/E_{\rm add}$ between the localized orbitals and the conduction electrons ($E_{\rm add}$ is a

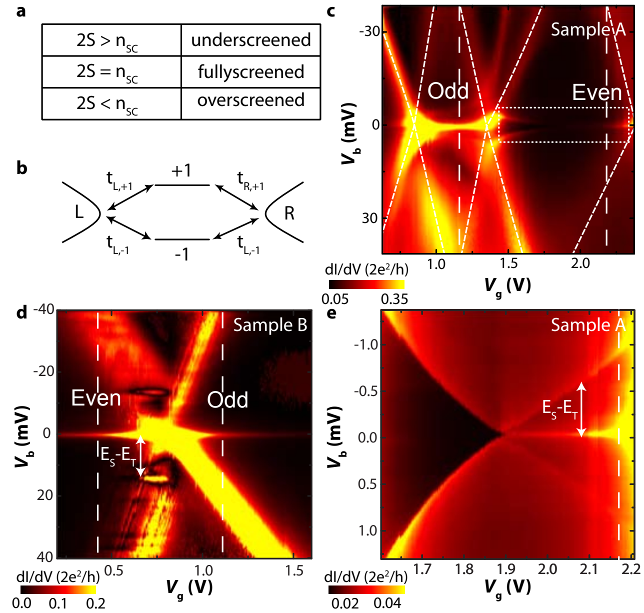

FIG. 1: **a.** Summary of the different types of Kondo effects according to impurity spin S and number of screening channels $n_{\rm sc}$; **b.** Tunneling model of our single molecule transistor: two orbital levels couple to two leads; **c.** Conductance map of sample A recorded at $T = 35$mK. Dashed lines are positioned at the gate voltages where more detailed studies where performed; **d.** Conductance map for sample B; **e.** Zoom inside the dotted rectangle defined in the even diamond of sample A. The white arrows represents the singlet-triplet splitting in both samples.

measure of the addition energy on the quantum dot). One then obtains two Kondo temperatures:

$$k_B T_{K\lambda} = \sqrt{D J_\lambda} e^{-1/(\rho J_\lambda)} \qquad (2)$$

where D is the bandwidth of the metal and ρ its density of states. While it seems physically sound that at vanishing temperature a spin $S = 1$ will be fully screened [20], moderate left/right asymmetries in the tunneling amplitudes (1) can lead to vastly different Kondo scales, because of the exponential behavior in (2), as pointed in [18]. This situation naturally occurs with electromigration, as the molecule tends to stay preferably closer to one of the electrodes, so that *e.g.* $t_{L,\pm1} \gg t_{R,\pm1}$. Fig. 1 shows indeed that our conductance maxima, here shown for two different devices, are much lower than the quantum value $2e^2/h$, with e the electron charge and h Planck's constant. It is then possible to have a large range of temperatures over which underscreened behavior prevails. A further crucial condition for the realization of the underscreened Kondo effect is the formation of a spin $S = 1$ unit itself. This relies on a strong Hund's rule that makes the separation between singlet and triplet states much larger than the temperature. These high-energy singlet excitations [22] are clearly spectroscopically resolved in all our measurements through cotunneling lines, see figure 1.d-e. Sample A in figure 1.c shows an odd charge Coulomb diamond with a pronounced Kondo ridge for gate voltages between V_g=0.8V and 1.35V (parity was argued in [19]), so that the next diamond seen up to V_g=2.3V has an even number of electrons. Figure 1.d represents the conductance map of sample B, where a single charge degeneracy point is observed, with Kondo ridges on *both* sides. We estimate a low limit for the addition energy $E_{\rm add} \gtrsim$200meV. Figure 1.e focuses inside the even diamond of sample A, where the singlet to triplet quantum phase transition occurs [19]. We can thus assess that our experiment was carried out in the temperature range $T_{K1} \ll T \ll T_{K2}$. Similar singlet excitations are visible on the left side of sample B, see Fig. 1.d., therefore allowing us to label this region even.

We now present the study of our two different devices (see figure 1.c-d), both showing fully screened and underscreened Kondo anomalies. In order to maximize the Kondo temperature, gate voltages were chosen away from the center of the Coulomb diamonds, while staying out of the mixed valence regime. In the fully screened Kondo effect, the conductance versus lowering the temperature has a logarithmic increase above the Kondo temperature, and then saturates in a quadratic, Fermi-liquid like fashion. On the other hand, the underscreened Kondo effect is expected to show two distinct logarithmic behaviors [4, 6, 7, 18], *above and below* the highest Kondo temperature T_{K2}. Indeed, partially screened moments are known to act as a ferromagnetic Kondo impurities [4], leading to a slow logarithmic scattering of the conduction electrons at low energy. This is best seen by plotting the derivative of conductance with respect to temperature as a function of inverse temperature, as suggested in [18]. Our data in the inset of figure 2a, taken for the even diamond of sample A, clearly display two *distinct* $1/T$ regimes. Another way to discriminate both Kondo effects is to perform a fit to NRG results [3, 23] of single channel spin $S = 1/2$ and $S = 1$ Kondo models. The obvious qualitative differences, namely the apparent divergence in $dG(T)/dT$ at low temperature T for spin $S = 1$ and the presence of a maximum for spin $S = 1/2$, in addition to the quantitative agreement with the NRG predictions are strong indications of two remarkably different Kondo states, see Fig. 2a-b. This comparison to theory allows us to extract the associated Kondo temperatures T_K and the conductance amplitude G_o for each curve, using the scaling form

$$G(T) = G_o f_S^{\rm NRG}(T/T_K) + G_{bg} \qquad (3)$$

with S the impurity spin and G_{bg} a constant conductance background [24]. We stopped our measurements around $T_{\rm base}$ =100mK in order to ensure that saturation effects are not provoked by an uncontrolled electronic temperature. We also limited the AC-excitation of the lock-in detection to stay in the limit $eV_{AC} \ll k_B T$. With these words of caution, we could not observe any saturation of the conductance over two decades of tempera-

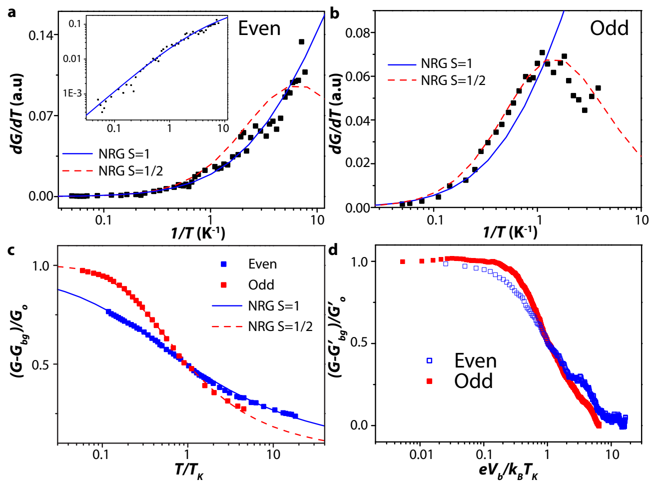

FIG. 2: **a.** Derivative of the conductance versus temperature recorded at gate $V_g = 2.17$V and bias $V_b = 0$V in the even Coulomb diamond of sample A (same data on a log scale in the inset, showing two distinct $1/T$ behaviors). The best fit is obtained for an underscreened Kondo model (blue) and gives $G_o = 0.14$ (in units of $2e^2/h$) and $T_K = 1.1$K; **b.** Derivative of the conductance versus temperature recorded at $V_g = 1.2$V and $V_b = 0$V in the odd Coulomb diamond. The best fit is obtained for a fully screened Kondo model (red) and gives $G_o = 0.34$ and $T_K = 4.4$K; **c.** Conductance data of a and b compared to the relevant NRG curves. The background was adjusted to follow the convention $G(T_K) = G_o/2$, giving $G_{bg} = 0.055$ in the fully screened case and $G_{bg} = 0.022$ in the underscreened case; **d.** Differential conductance at the base temperature rescaled in universal form, with $G'_o = G(T_{\mathrm{base}})$, and G'_{bg} such that the conductance is $G'_o/2$ at $eV_b = k_B T_K$.

ture in the even diamond, contrary to what is witnessed in the fully screened case. Since the Kondo effect displays an universal behavior with a single scale T_K, this must be reflected by different universal curves between screened and underscreened situations, as evidenced by Fig. 2c. The bias voltage dependence of the non-linear conductance at the base temperature was also examined [25, 26]. Although this study is more difficult to perform accurately, two relatively different curves are nevertheless obtained, with slower voltage behavior for the spin $S = 1$ molecules, see Fig. 2d. We also note that a measure of T_K can be roughly deduced from the half width at half maximum (HWHM) of the non-linear conductance peak, and this method was used to have an alternative determination of T_K for each Kondo ridge in sample A and B, after extraction of a background contribution (see table I). The higher error bars for the bias method are due to the absence of reliable finite voltage predictions from theory, so that T_K depends sensitively on the estimated background. Both methods, relying on thermal or voltage smearing of the Kondo peak, nicely converge to comparable T_K values for *both* types of Kondo effects.

Sample/Method	Temperature	Bias	Magnetic Field
A (odd)	$4.4 \pm 0.3K$	$5.5 \pm 1.3K$	$4.8 \pm 0.3K$
A (even)	$1.1 \pm 0.1K$	$1.9 \pm 0.5K$	$0.6 \pm 0.4K$
B (odd)	none	$4.4 \pm 0.8K$	$5.4 \pm 0.3K$
B (even)	none	$1.9 \pm 0.3K$	$0.2 \pm 0.1K$

TABLE I: Kondo temperatures for each sample in the even and odd diamonds, determined by using the methods of temperature, bias or magnetic field, as described in the text. Sample B was not stable enough to perform a detailed temperature study.

The evolution of the Kondo peak as a function of magnetic field, as seen in the bias spectrum of Fig. 3.a-b, displays even more dramatic effects. Previous experimental studies [17, 27, 28] and theoretical calculations [29] on fully screened impurities have shown a Zeeman splitting of the Kondo resonance at a critical field $g\mu_B B_c \sim 0.5 k_B T_K$. Indeed, as seen in table I, this estimate agrees with the T_K determined by the temperature and bias dependence, for the odd charge diamonds of both samples. In contrast, *for the spin $S = 1$ quantum dot, the Kondo resonance separates into Zeeman states at a much lower magnetic field*, $g\mu_B B_c \ll k_B T_K$, so that the Kondo temperature naively determined from $2g\mu_B B_c$ is in disagreement with the previous estimates, see table I. This observation can be understood as the combination of two related effects. First, a partially screened Kondo impurity is expected to be highly polarizable in a small magnetic field, which constitutes a relevant perturbation to the free spin fixed point, in the renormalization group sense. Second, the logarithmic cusp of the Kondo peak at low bias makes the splitting more pronounced than for the Lorentzian shape typical of a Fermi Liquid in the fully screened case. To put this surprising observation on firmer ground, we have performed NRG calculations for the local density of states in a magnetic field for the single-channel Kondo models with spins $S = 1/2$ and $S = 1$. This numerical solution allows us to confirm that the underscreened Kondo resonance starts to unveil its magnetic excitations for Zeeman energies as low as $k_B T_K/16$, see figure 3.d-e. A further interesting check on our analysis lies on the extracted Zeeman splitting as a function of magnetic field, figure 3.c. For odd/even diamonds respectively, we find that the dispersion of the magnetic states, rescaled to the corresponding Kondo temperature, clearly shows the presence/absence of a threshold. Also remarkable is how these data for our two *different* samples quite naturally fall on top of each other, another signature of Kondo universality.

In conclusion, we have given comprehensive experimental evidence for the occurence of Nozières and Blandin underscreened Kondo effect in even charge molecular quantum dots. Our analysis, based on the

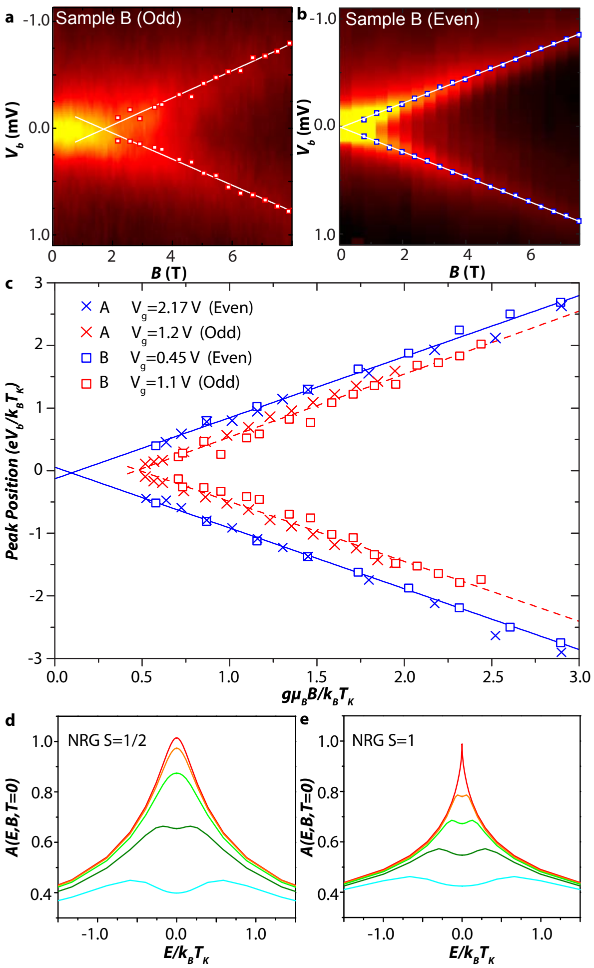

FIG. 3: **a. b.** Differential conductance versus bias voltage and magnetic field for sample B in the odd and even diamonds (lines are linear fits to the B-dependence of the Zeeman peaks of sample B); **c.** Positions of the Zeeman peaks of both samples and odd/even diamonds, as extracted from above. Linear fits provide the critical field B_c by extrapolation to zero bias. Both bias voltage and magnetic field were renormalized by T_K deduced from the HWHM at $B = 0$T for each case; **d. e.** NRG calculations of the energy E dependent equilibrium local density of states $A(E, B, T = 0)$ at zero temperature, normalized to its $E = B = 0$ value, for the spin $S = 1/2$ and $S = 1$ single-channel Kondo models and magnetic field values $g\mu_B B/k_B T_K = 0, 1/8, 1/4, 1/2, 1$ (top to bottom).

stark differences with respect to regular transport properties of fully screened impurities, was strenghtened by NRG calculations. An unexpected magnetic field sensitivity of partially screened Kondo impurities was also discovered, that we could confirm theoretically. This work illustrates the striking versatility of molecular electronics for the investigation of fundamental aspects in quantum magnetism.

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SUPPLEMENTARY INFORMATION ON "OBSERVATION OF THE UNDERSCREENED KONDO EFFECT IN A MOLECULAR TRANSISTOR"

On the Kondo scaling of the temperature-dependent conductance

A hallmark of Kondo dominated quantum transport in nanostructures is the universal one-parameter scaling of the linear conductance as a function of the ratio of temperature T to the Kondo scale T_K:

$$G(T) = G_o\, f(T/T_K) + G_{bg} \tag{1}$$

with G_0 the overall magnitude of the conductance variation (related to the asymmetry in lead couplings and the total hybridization Γ to the leads), G_{bg} a high temperature background associated with direct tunneling processes, and $f(x)$ an universal scaling function that depends solely on the spin S carried by the quantum dot. An important feature of this physical quantity is the broad crossover taking place between the two extreme limits of temperature, $T \ll T_K$ and $T \gg T_K$. For the spin $S = 1/2$ fully screened case, the scaling function behaves as $f(x) = 1 - (\pi^4/16)x^2$ and $f(x) = (3\pi^2/16)/\log^2(x)$ in the Fermi liquid ($x \ll 1$) and local moment ($x \gg 1$) regimes respectively [1]. The complete crossover curve, associated with the full function $f(x)$ for all x, can be accurately calculated from Wilson's NRG [2]

In practice, however, the universal high temperature regime is attained in experiment only if the Kondo scale T_K happens to be at least two orders of magnitude smaller than the Coulomb repulsion U on the quantum dot [3]. In that case, Coulomb blockade effects have already set in at temperatures much higher than T_K, ensuring that one can safely extract $f(x)$ for $x \gg 1$, corresponding to the logarithmic Kondo tails. In terms of the microscopic Anderson model, this condition corresponds to $U/\pi\Gamma > 2$, where the universal crossover from the local moment to the Fermi liquid regimes takes place (the Wilson ratio is then quite close to 2) [3]. This clearly sets a first important constraint on the value of the charging energy when analyzing an experiment in terms of universal Kondo behavior. Now, to assess experimentally the low temperatures regime, associated with the behavior of $f(x)$ at $x \ll 1$, it is crucial that the actual Kondo temperature is not vanishingly small, preferably an order of magnitude higher than the base temperature $T_{\rm base}$ of the cryostat. When the above two conditions, $10T_{\rm base} < T_K < U/100$, are met together, the conductance shows a characteristic inverted-S shape, see Fig.1a, that provides a very strong constraint to the fitting procedure with the NRG calculation. In this case, all non-universal parameters (T_K, G_o, G_{bg}) can be accurately extracted from the data. On the contrary, when the Kondo scale does not obey one of these two requirements, the incomplete determination of the crossover function makes the fit quite sensitive to the estimation of the background G_{bg}, resulting in an imprecise determination of T_K. In our experiment the Coulomb energy U is of the order of 1000K, the Kondo scale T_K is 4K (for the spin $S = 1/2$ samples), and our temperature ranges from 100mK to 20K, so that we can clearly capture the *whole* Kondo crossover, and determine T_K unambiguously. The full development of the Kondo resonance is similarly observed in the non-linear conductance, Fig.1b, which becomes effectively very broad in the high temperature regime, while saturating to a constant height and width below 200mK.

The method to address the underscreened Kondo effect in our integer spin samples follows exactly the same strategy of fitting the complete crossover function to the universal result from NRG calculations on the spin $S = 1$ Kondo model. Here, the Kondo scale T_K is found to be close to $1K$ at the gate voltage $V_g = 2.17V$ for sample A, using a quantitative fit to the NRG result as shown in Fig.2. Again, this conductance displays both the Kondo tails (for $T > T_K$) and a slightly different behavior (for $T < T_K$). The latter clearly appears to be much slower than the clear saturation measured in the spin $S = 1/2$ samples (see Fig.1a) and is associated with the second logarithmic regime *below* the Kondo scale, which is a characteristic "smoking gun" signature of the underscreened Kondo effect. This interpretation is further strengthened by the quantitative agreement to the NRG calculations. Measurements at two further gate voltages have also been taken, and data collapse onto the universal curve for the underscreened Kondo conductance is reasonably good, see Fig.2. We stress, however, that these two extra measurements, performed nearer to the center of the Coulomb diamond, are associated with relatively smaller Kondo temperatures in the 200mK range, and do not allow to probe the *whole* Kondo crossover from well below to well above T_K, in contrast to our measurement at the optimal gate voltage at 2.17V. The key to maximizing T_K is thus to benefit from the virtual charge fluctuations on the side of the Coulomb diamond, while staying away from the mixed valence regime, leading to an optimal choice of voltage where the experiment should be performed.

Zeeman effect: threshold behavior in the non-linear conductance

A second key signature of the underscreened Kondo effect (with respect to the standard fully screened case) lies in an anomalous Zeeman effect of the Kondo resonance, as found in our study. For completeness, we present here the raw non-linear conductance traces, that display the same data, yet in a different fashion than the color plots

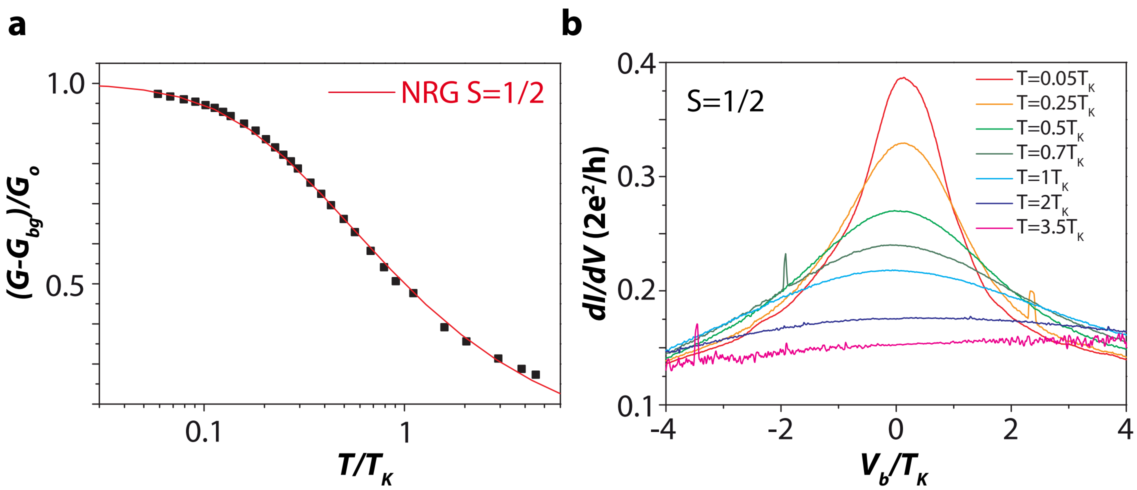

FIG. 1: **a.** Linear conductance as a function of temperature for sample A in the half-integer spin region (gate voltage $V_g = 1.2$V), rescaled in universal form, and compared to NRG calculations on the spin $S = 1/2$ Kondo model. **b.** Related non-linear conductance traces as a function of bias (rescaled by the Kondo scale T_K) for several different temperatures.

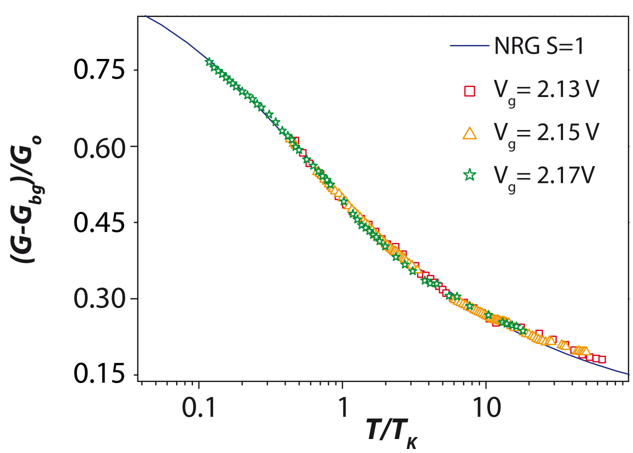

FIG. 2: Linear conductance as a function of temperature for sample A in the integer spin region (gate voltages $V_g = 2.13, 2.15, 2.17$V), rescaled in universal form, and compared to NRG calculations on the spin $S = 1$ Kondo model.

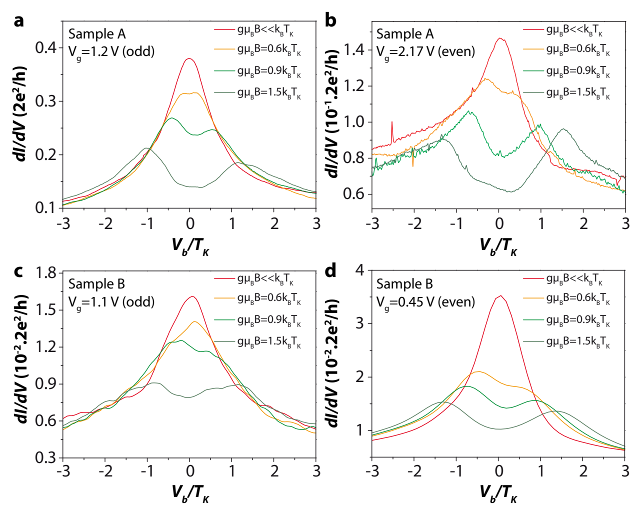

FIG. 3: Non-linear conductance traces as a function of bias (rescaled by the Kondo scale T_K) for fixed values of the ratio of magnetic field B to the Kondo temperature T_K. Regions associated to spin $S = 1/2$ and $S = 1$ for sample A, taken at the base temperature, are shown in panels **a** and **b** respectively, which corresponds to the linear conductances given in Figs.1a and 2. Similar data for sample B are shown in panels **c** and **d**, corresponding to odd and even Coulomb diamonds respectively.

given in the main text. Here again, odd and even charge diamonds for two different samples are investigated at "optimal" gate voltages (see discussion above), and the conductance traces at the base temperature for several values of the magnetic field are shown in Fig.3. All data are rescaled by the corresponding Kondo temperature T_K, both with respect to bias voltage V_b and magnetic field B. It is clearly apparent that the Zeeman splitting in the Kondo peak occurs at lower values of B/T_K in the case of spin $S = 1$ samples. This phenomenon reflects the high degree of polarizability of the underscreened Kondo impurity, and this anomalous Zeeman effect constitutes a crucial fingerprint of the underscreened Kondo effect, that can be used as a guide for further experimental investigations.

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